Magnetic free energy at elevated temperatures and hysteresis of magnetic particles

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Abstract

We derive a free energy for weakly anisotropic ferromagnets which is valid in the whole range of temperature and interpolates between the micromagnetic energy at zero temperature and the Landau free energy near the Curie point T_c. This free energy takes into account the change of the magnetization length due to thermal effects, in particular, in the inhomogeneous states. As an illustration, we study the thermal effect on the Stoner-Wohlfarth curve and hysteresis loop of a ferromagnetic nanoparticle assuming that it is in a single-domain state. Within this model, the saddle point of the particle's free energy, as well as the metastability boundary, are due to the change in the magnetization length sufficiently close to T_c, as opposed to the usual homogeneous rotation process at lower temperatures.

1 Introduction

The macroscopic energy of weakly anisotropic ferromagnets which first appeared in the seminal paper by Landau and Lifshitz [1], has been an instrument for innumerable investigations of domain walls and other inhomogeneous states of magnetic systems. Later an approach based on this macroscopic energy has been called "micromagnetics" [2]. Strictly speaking, micromagnetics is an essentially zero-temperature theory for *classical* magnets, as it considers the magnetization as a vector of fixed length. Under these conditions, it can be easily obtained as a continuous limit of the classical Hamiltonian on a lattice. Practically, micromagnetics has been applied to nonzero temperatures as well, with temperature-dependent equilibrium magnetization and anisotropy constants.

On the other hand, close to the Curie temperature T_c a magnetic free energy of the Landau type can be considered. This free energy allows the magnetization

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to change both in direction and length, and it is, in fact, a continuous limit of the free energy following from the mean-field approximation (MFA). Using this approach, Bulaevskii and Ginzburg [3] predicted a phase transition between the Bloch walls and the Ising-like walls at temperatures slightly below T_c. Much later this phase transition was observed experimentally [4, 5] using the theoretical results for the mobility of domain walls in this regime [6, 7].

Although both of these approaches have been formulated at the same mean-field level, they have been considered as unrelated for a long time. On the other hand, the MFA itself is an all-temperature approximation, and thus one can question its macroscopic limit in the whole temperature range. The corresponding macroscopic free energy should interpolate between the Landau-Lifshitz energy, or micromagnetics at $T = 0$ and the Landau free energy in the vicinity of T_c. The Euler equation for the magnetization which minimizes this generalized free energy appears in Refs. [6] and [8]. Whereas the condition for the Landau theory is $M \ll M_s$, where M_s is the saturation magnetization at $T = 0$, the new equations only require $|M - M_e| \ll M_s$, where M_e is the equilibrium magnetization at zero field. For weakly anisotropic magnets (the anisotropy energy is much less than the homogeneous exchange energy, and this is satisfied by most compounds) the magnetization magnitude M is either small or only slightly deviates from M_e, thus the condition above is satisfied in the whole temperature range.

Derivation of the generalized macroscopic free energy from the MFA is much subtler than that of the Euler equations. This free energy appears in Ref. [8] where its form was guessed. In this paper, we will present this derivation and illustrate the resulting magnetic free energy in the case of a single-domain magnetic particle with a uniaxial anisotropy. For the sake of transparency, we will consider the exchange anisotropy rather than the single-site anisotropy; this does not change the qualitative results. It will be shown that the free-energy landscape for the magnetic particle has different forms at low temperatures and near T_c. At low temperatures, the saddle point between the "up" and "down" minima is located near the sphere $|\mathbf{M}| = M_e$, the value of M being somewhat reduced in comparison with M_e due to thermal effects at $T > 0$. Near T_c, the value of M strongly changes by going from one minimum to another through the saddle point; for zero fields the saddle point becomes $\mathbf{M} = \mathbf{0}$. These effects also modify the metastability boundary of the magnetic particle (the famous Stoner-Wohlfarth curve [9] which was experimentally observed in Ref. [10]), and its hysteresis curves.

The main body of this paper is organized as follows. In Sec. 2 we give the derivation of the magnetic free energy at all temperatures. In Sec. 3 the free-energy landscape of a single-domain magnetic particle is analyzed. In Secs. 4 and 5 we study the Stoner-Wohlfarth curves and hysteresis loops at different temperatures. In Sec. 6 we discuss the possibility of observing the new thermal effects for single-domain magnetic particles.

2 Free energy of a magnetic particle

Let us start with the *biaxial* ferromagnetic model described by the classical anisotropic Hamiltonian of the type

$$\mathcal{H} = -\mu_0 \mathbf{H} \sum_i \mathbf{s}_i - \frac{1}{2} \sum_{ij} J_{ij}(s_{zi}s_{zj} + \eta_y s_{yi}s_{yj} + \eta_x s_{xi}s_{xj}), \tag{1}$$

where μ_0 is the magnetic moment of the atom, i, j are lattice sites, $\mathbf{s}_i$ is the normalised vector, $|\mathbf{s}_i| = 1$, and the dimensionless anisotropy factors satisfy $\eta_x \leq \eta_y \leq 1$.

To study the macroscopic properties of this system at nonzero temperatures, it is convenient to use a macroscopic free energy. In the literature one can find two types of macroscopic free energies for magnets. One of them is the so-called micromagnetic free energy which is valid at zero temperature, and the other one is Landau's free energy which is applied near the Curie temperature T_c. Since both free energies are based on the mean-field approximation (MFA), it is possible to derive a simple form of the MFA free energy for weakly anisotropic ferromagnets which is valid in the whole temperature range and bridges these two well-known forms.

The free energy $F = -T \ln \mathcal{Z}$ of a spin system described by the Hamiltonian in Eq. (1) can be calculated in the mean-field approximation by considering each spin on a site i as an isolated spin in the effective field containing contributions determined by the mean values of the neighboring ones. Namely,

$$\mathcal{H} \Rightarrow \mathcal{H}^{\mathrm{MFA}} = \mathcal{H}_{00} - \sum_i \mathbf{H}_i^{\mathrm{MFA}} \mathbf{s}_i, \tag{2}$$

where

$$\mathcal{H}_{00} = \frac{1}{2} \sum_{ij} J_{ij} \left(\sigma_{zi}\sigma_{zj} + \eta_x \sigma_{xi}\sigma_{xj} + \eta_y \sigma_{yi}\sigma_{yj} \right), \tag{3}$$

$\boldsymbol{\sigma}_i \equiv \langle \mathbf{s}_i \rangle$ is the spin polarization, and the molecular field $\mathbf{H}_i^{\mathrm{MFA}}$ is given by

$$\mathbf{H}_i^{\mathrm{MFA}} = \mu_0 \mathbf{H} + \sum_j J_{ij} \left(\sigma_{zj}\mathbf{e}_z + \eta_x \sigma_{xj}\mathbf{e}_x + \eta_y \sigma_{yj}\mathbf{e}_y \right). \tag{4}$$

Then the solution of the one-spin problem in Eq. (2) leads to

$$F = \mathcal{H}_{00} - NT \ln(4\pi) - T \sum_i \Lambda(\xi_i)$$

$$\Lambda(\xi) \equiv \ln \left(\frac{\sinh(\xi)}{\xi} \right), \tag{5}$$

where N is the total number of spins, $\xi_i \equiv |\boldsymbol{\xi}_i|$, and $\boldsymbol{\xi}_i \equiv \beta \mathbf{H}_i^{\mathrm{MFA}}$. The MFA free energy determined by Eq. (4) and Eq. (5) can be minimized with respect to the spin averages $\boldsymbol{\sigma}_i$ to find the equilibrium solution in the general case where the anisotropy $1 - \eta_{x,y}$ is not necessarily small. The minimum condition for the free energy, $\partial F/\partial \boldsymbol{\sigma}_i = 0$, leads to the Curie-Weiss equation

$$\boldsymbol{\sigma}_i = B(\xi_i) \frac{\boldsymbol{\xi}_i}{\xi_i}, \tag{6}$$

where $B(\xi) = \coth(\xi) - 1/\xi$ is the Langevin function.

For small anisotropy, $1 - \eta_{x,y} \ll 1$, one can go over to the continuum limit and write for the short-range interaction J_{ij}

$$\sum_j J_{ij}\boldsymbol{\sigma}_j \cong J_0\boldsymbol{\sigma}_i + J_0\alpha\Delta\boldsymbol{\sigma}_i, \tag{7}$$

where Δ is the Laplace operator acting on the components of $\boldsymbol{\sigma}(\mathbf{r})$, J_0 is the zero Fourier component (the zeroth moment), and $J_0\alpha$ is the second moment of the exchange interaction J_{ij}. For the simple cubic lattice with nearest neighbor interactions $\alpha = a_0^2/z$, $z = 6$, and a_0 is the lattice spacing. Going from summation to integration in Eq. (5), one obtains

$$\frac{F}{J_0} = -\frac{NT}{J_0}\ln(4\pi) + \frac{1}{v_0}\!\int\! d\mathbf{r}\left\{\frac{1}{2}\sigma^2 + \frac{1}{2}(\boldsymbol{\sigma}, \mathbf{h}_{\text{eff}} - \mathbf{h}) - \frac{1}{\beta J_0}\Lambda(\xi)\right\}, \tag{8}$$

where v_0 is the unit-cell volume,

$$\begin{aligned}\boldsymbol{\xi} &= \beta J_0(\boldsymbol{\sigma} + \mathbf{h}_{\text{eff}}) \\ \mathbf{h}_{\text{eff}} &= \mathbf{h} + \alpha\Delta\boldsymbol{\sigma} - (1 - \eta_x)\sigma_x\mathbf{e}_x - (1 - \eta_y)\sigma_y\mathbf{e}_y,\end{aligned} \tag{9}$$

and $\mathbf{h} \equiv \mu_0\mathbf{H}/J_0$. We will consider the case of small fields, $h \ll 1$. Since in this case in Eq. (9) $|\mathbf{h}_{\text{eff}}| \ll |\boldsymbol{\sigma}|$ in the whole range below T_c (near T_c the value of σ is small but the susceptibility is large), the last term of Eq. (8) can be expanded to first order in $\mathbf{h}_{\text{eff}}$ using

$$\xi = \xi_0 + \delta\xi, \qquad \xi_0 = \beta J_0\sigma, \qquad \delta\xi \cong \beta J_0\frac{\boldsymbol{\sigma}\mathbf{h}_{\text{eff}}}{\sigma} \tag{10}$$

and the first two terms of the expansion

$$\Lambda(\xi) \cong \Lambda(\xi_0) + B(\xi_0)\delta\xi + \frac{1}{2}B'(\xi_0)(\delta\xi)^2, \tag{11}$$

where $B'(\xi) \equiv dB(\xi)/d\xi$. Hence,

$$\begin{aligned}\frac{F}{J_0} &= \frac{1}{v_0}\!\int\! d\mathbf{r}\left\{\frac{1}{2}\sigma^2 - \frac{1}{\beta J_0}\Lambda(\xi_0) - \frac{B(\xi_0)}{\sigma}\boldsymbol{\sigma}\mathbf{h} - \left(\frac{B(\xi_0)}{\sigma} - \frac{1}{2}\right)(\boldsymbol{\sigma}, \mathbf{h}_{\text{eff}} - \mathbf{h})\right\} \\ &\quad - \frac{NT}{J_0}\ln(4\pi).\end{aligned} \tag{12}$$

Near $T_c = T_c^{\text{MFA}} = J_0/3$ the order parameter $\boldsymbol{\sigma}$ becomes small, and using $\Lambda(\xi) \cong \xi^2/6 - \xi^4/180$ and $B(\xi_0) \cong \xi_0/3 \cong \beta_c J_0\sigma/3 = \sigma$ one straightforwardly arrives at the Landau free energy

$$\begin{aligned}\frac{F}{J_0} &= \frac{1}{v_0}\!\int\! d\mathbf{r}\left\{\frac{1}{2}\alpha(\nabla\boldsymbol{\sigma})^2 - \boldsymbol{\sigma}\mathbf{h} + \frac{1}{2}(1 - \eta_x)\sigma_x^2 + \frac{1}{2}(1 - \eta_y)\sigma_y^2 - \frac{\epsilon}{2}\sigma^2 + \frac{3}{20}\sigma^4\right\} \\ &\quad - \frac{NT}{J_0}\ln(4\pi),\end{aligned} \tag{13}$$

where $\epsilon \equiv (T_c^{\text{MFA}} - T)/T_c^{\text{MFA}}$ and $(\nabla\sigma)^2 = (\nabla\sigma_x)^2 + (\nabla\sigma_y)^2 + (\nabla\sigma_z)^2$. Note that Eq. (13) formally yields unlimitedly increasing values of σ at equilibrium,

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as a function of the field h. To comply with the condition $\sigma \ll 1$ in Landau's formalism, h should be kept small, as was required above. One could also work out the h^2 corrections to Eq. (13).

Now we consider the temperature region where the influence of anisotropy and field on the magnitude of the spin polarization σ can be studied perturbatively. Concerning the influence of anisotropy, the applicability criterion can be obtained from the requirement that the local shift of T_c for spins forced perpendicularly to the easy axis should be smaller than the distance from T_c, i.e., $\Delta T_c / T_c \sim 1 - \eta \ll \epsilon$. The macroscopic free energy in the perturbative region can then be combined with the Landau free energy in their common applicability range $1 - \eta \ll \epsilon \ll 1$. Thus, in the perturbative region we expand the first two terms of expression Eq. (12) up to the second order in $\delta\sigma \equiv \sigma - \sigma_e$ using

$$\xi_0 = \xi_e + \delta\xi, \qquad \xi_e = \beta J_0 \sigma_e, \qquad \delta\xi \cong \beta J_0 \delta\sigma \qquad (14)$$

and the formula analogous to Eq. (11). This leads to

$$\frac{F}{J_0} = \frac{F_e}{J_0} + \frac{1}{v_0} \int d\mathbf{r} \left\{ \frac{1}{2}(1 - B'\beta J_0)(\sigma - \sigma_e)^2 - \frac{B(\xi_0)}{\sigma}\boldsymbol{\sigma}\mathbf{h} - \left(\frac{B(\xi_0)}{\sigma} - \frac{1}{2} \right)(\boldsymbol{\sigma}, \mathbf{h}_{\text{eff}} - \mathbf{h}) \right\} , (15)$$

where $B' = B'(\xi_e)$, and

$$\frac{F_e}{J_0} = -\frac{NT}{J_0}\ln(4\pi) + N\left[\frac{1}{2}\sigma_e^2 - \frac{1}{\beta J_0}\Lambda(\xi_e) \right], \qquad (16)$$

is the equilibrium free energy in the absence of magnetic field and the quantity σ_e is the spin polarisation at equilibrium satisfying the homogeneous Curie-Weiss equation

$$\sigma_e = B(\xi_e). \qquad (17)$$

The minimum condition for Eq. (15), $\delta F/\delta\boldsymbol{\sigma} = 0$, after an accurate calculation taking into account the dependence of $B(\xi_0)$ on σ and neglecting the terms quadratic in $\mathbf{h}_{\text{eff}}$, results in an equation of the form

$$\frac{1}{\bar{\chi}_{\parallel}}(\sigma - \sigma_e)\frac{\boldsymbol{\sigma}}{\sigma} - \mathbf{h}_{\text{eff}} + \frac{[\boldsymbol{\sigma} \times [\boldsymbol{\sigma} \times \mathbf{h}_{\text{eff}}]]}{\sigma^2 \bar{\chi}_{\parallel}} = 0, \qquad (18)$$

where $\mathbf{h}_{\text{eff}}$ is given by Eq. (9) and the dimensionless longitudinal susceptibility $\bar{\chi}_{\parallel}$ is given in Eq. (19) below. The solution of Eq. (18) satisfies $\boldsymbol{\sigma} \| \mathbf{h}_{\text{eff}}$, and the term with the double vector product plays no role. Considering the response to small fields $\mathbf{h} = h_z\mathbf{e}_z$ and $\mathbf{h} = h_{x,y}\mathbf{e}_{x,y}$ in Eq. (18) in a homogeneous situation (in the transverse case $\mathbf{h}_{\text{eff}} = 0$ and $\sigma = \sigma_e$), one can identify the reduced susceptibilities for the spin polarization as

$$\bar{\chi}_{\parallel} \equiv \frac{d\sigma_z}{dh_z} = \frac{B'\beta J_0}{1 - B'\beta J_0},$$

$$\bar{\chi}_x = \frac{1}{1 - \eta_x}, \qquad \bar{\chi}_y = \frac{1}{1 - \eta_y}. \qquad (19)$$

Our expression for the free energy, Eq. (15), is still cumbersome, but can be simplified if we make the observation that in the perturbative region the deviation

$\delta\sigma \equiv \sigma - \sigma_e$ is proportional to h_{eff}, and the terms of the type $\delta\sigma \cdot h_{\text{eff}}$ and $(\delta\sigma)^2$ in Eq. (15) are thus quadratic in h_{eff}. Such terms are nonessential in the calculation of F itself, they are only needed for the proper writing of the equilibrium equation Eq. (18). Now we can replace Eq. (15) by a simplified form

$$\frac{F}{J_0} = \frac{F_e}{J_0} + \frac{1}{v_0}\int d\mathbf{r} \left\{ \frac{1}{2}\alpha(\nabla\boldsymbol{\sigma})^2 - \boldsymbol{\sigma}\mathbf{h} + \frac{1}{2}(1-\eta_x)\sigma_x^2 + \frac{1}{2}(1-\eta_y)\sigma_y^2 + \frac{1}{2\bar{\chi}_\parallel}(\sigma-\sigma_e)^2 \right\} . (20)$$

This form coincides with Eq. (15), if we set $\sigma = \sigma_e$, i.e., it yields the same value of F in the leading first order in h_{eff}. On the other hand, Eq. (20) leads to the same equilibrium equation, Eq. (18), without the nonessential last term. Now, at the last step of the derivation, one can combine Eq. (20) with the Landau free energy in Eq. (13), leading to

$$\frac{F}{J_0} = \frac{F_e}{J_0} + \frac{1}{v_0}\int d\mathbf{r} \left\{ \frac{1}{2}\alpha(\nabla\boldsymbol{\sigma})^2 - \boldsymbol{\sigma}\mathbf{h} + \frac{1}{2\bar{\chi}_x}\sigma_x^2 + \frac{1}{2\bar{\chi}_y}\sigma_y^2 + \frac{1}{8\sigma_e^2\bar{\chi}_\parallel}(\sigma^2-\sigma_e^2)^2 \right\} . (21)$$

Indeed, in the Landau region, $\epsilon \ll 1$, from Eq. (17) and Eq. (19) it follows $\sigma_e^2 \cong (5/3)\epsilon$ and $\bar{\chi}_\parallel \cong (2\epsilon)^{-1}$, and Eq. (21) simplifies to Eq. (13). In terms of the magnetization $\mathbf{M}$ defined by

$$\mathbf{M}(\mathbf{r}) = \mu_0\boldsymbol{\sigma}(\mathbf{r})/v_0, \qquad \boldsymbol{\sigma}(\mathbf{r}) \equiv \langle\mathbf{s_r}\rangle, \tag{22}$$

and other dimensional quantities, the free energy Eq. (21) takes on the form [5, 8]

$$F = F_e + \int d\mathbf{r} \left\{ \frac{1}{2q_d^2}(\nabla\mathbf{M})^2 - \mathbf{M}\cdot\mathbf{H} + \frac{1}{2\chi_x}M_x^2 + \frac{1}{2\chi_y}M_y^2 + \frac{1}{8M_e^2\chi_\parallel}(M^2-M_e^2)^2 \right\} , (23)$$

with

$$q_d^2 = \frac{W_D}{\alpha J_0}, \qquad \chi_\alpha = \frac{W_D}{J_0}\bar{\chi}_\alpha, \qquad W_D \equiv \frac{(\mu_0)^2}{v_0}, \tag{24}$$

where q_d is the so-called dipolar wave number, W_D is the characteristic energy of the dipole-dipole interaction, and $\chi_\alpha \equiv dM_\alpha/dH_\alpha$ with $\alpha = x, y, z$ are the susceptibilities [cf. Eq. (19)]. The applicability of Eq. (23) requires that the deviation $M - M_e$ from the equilibrium magnetization M_e in the absence of anisotropy and field, is small in comparison with the saturation value $M_s = \mu_0/v_0$. This is satisfied in the whole range of temperature if the anisotropy and field are small, i.e., $1 - \eta_{x,y} \ll 1$ and $\mu_0 H \ll J_0$. On the other hand, the free energy in Eq. (23) can be transformed into the "micromagnetic" form by introducing the magnetization direction vector $\boldsymbol{\nu} \equiv \mathbf{M}/M$. One can then write

$$\frac{1}{2\chi_{x,y}}M_{x,y}^2 = K_{x,y}\nu_{x,y}^2, \qquad K_{x,y} = \frac{M^2}{2\chi_{x,y}}, \tag{25}$$

where $K_{x,y}$ are the anisotropy constants. In particular, for the uniaxial model one can rewrite

$$\frac{1}{2\chi_\perp}(M_x^2 + M_y^2) = -K\nu_z^2 + K, \tag{26}$$

where K is the uniaxial anisotropy constant. Note that at nonzero temperatures M and thus the anisotropy constants can be spatially inhomogeneous. In this case Eq. (23) is more useful than its micromagnetic form.

It should be stressed that the traditional way of writing the magnetic free energy in terms of the magnetization is, at least from the theoretical point of view, somewhat artificial. All terms in Eq. (23) apart from the Zeeman term, are non-magnetic and in fact independent of the atomic magnetic moment μ_0. The latter cancels out of the resulting formulae, as soon as they are reexpressed in terms of the original Hamiltonian (1). On the other hand, Eq. (23) is convenient if the parameters are taken from experiments.

Examination of the formalism above shows that it can be easily generalized to quantum systems, leading to the same form as in Eq. (23). In the derivation, the classical Langevin function $B(x)$ is replaced by the quantum Brillouin function $B_S(x)$.

3   The free-energy landscape for a uniaxial magnetic particle

Henceforth, we will consider the uniaxial anisotropy, $\eta_x = \eta_y \equiv \eta_\perp$. For single-domain magnetic particles in a homogeneous state, the gradient terms in the free energy can be dropped and the free energy of Eq. (23) can be presented in the form

$$F = F_e + (V M_e^2 / \chi_\perp) f$$
$$f = -\mathbf{n} \cdot \mathbf{h} + \frac{1}{2}(n_x^2 + n_y^2) + \frac{1}{4a}(n^2 - 1)^2, \tag{27}$$

where V is the particle's volume and f the reduced free energy written in terms of the reduced variables

$$\mathbf{n} \equiv \mathbf{M}/M_e, \qquad \mathbf{h} \equiv \mathbf{H}\chi_\perp/M_e, \qquad a \equiv 2\chi_\parallel/\chi_\perp. \tag{28}$$

One can see that the parameter a here controls the rigidity of the magnetization vector; it goes to zero in the zero-temperature limit (the fixed magnetization length) and diverges at T_c as $a \cong (1 - \eta)/\epsilon$ within the MFA. As the MFA is not quantitatively accurate, it is better to consider the susceptibilities and hence a as taken from experiments. Although this procedure is not rigorously justified, it can improve the results. Note that the reduced free energy f in Eq. (27) is only defined for $T < T_c$ since the reduced magnetization $\mathbf{n}$ is normalized by M_e.

For fields $\mathbf{h}$ inside the Stoner-Wohlfarth astroid, which will be generalized here to nonzero temperatures, f has two minima separated by a barrier. Owing to the axial symmetry, one can set $n_y = 0$ for the investigation of the free energy landscape. The minima, saddle points, and the maximum can be found from the equations $\partial f/\partial n_x = \partial f/\partial n_z = 0$, or, explicitly

$$n_z(n^2 - 1) = ah_z$$
$$n_x(n^2 - 1 + a) = ah_x. \tag{29}$$

These equations can be rewritten as

$$1 - n^2 = -ah_z/n_z = a(1 - h_x/n_x), \tag{30}$$

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whereupon the important relation follows

$$\frac{h_x}{n_x} - \frac{h_z}{n_z} = 1 \qquad \text{or} \qquad n_x = \frac{h_x n_z}{h_z + n_z}. \tag{31}$$

Using the latter, one can solve for n_x and obtain a 5th-order equation for n_z

$$h_x^2 n_z^3 = (h_z + n_z)^2 (ah_z + n_z - n_z^3), \tag{32}$$

from which the parameters of the potential landscape such as energy minima, saddle point, and energy barriers can be found.

In zero field, the characteristic points of the energy landscape can be simply found from Eqs. (29). One of these points is $n_x = n_z = 0$, which is a local maximum for $a < 1$ and a saddle point for $a > 1$. The minima are given by $n_x = 0$, $n_z = \pm 1$. The saddle points correspond to $n_z = 0$, while from the second of Eqs. (29) one finds

$$n_x = \begin{cases} \pm\sqrt{1-a}, & a \leq 1 \\ 0, & a \geq 1. \end{cases} \tag{33}$$

In fact, due to the axial symmetry, for $a < 1$ one has a saddle circle $n_x^2 + n_y^2 = 1 - a$ rather than two saddle points. The free-energy barrier following from this solution is given by

$$\Delta f \equiv f_{\text{sad}} - f_{\text{min}} = \begin{cases} (2-a)/4, & a \leq 1 \\ 1/(4a), & a \geq 1. \end{cases} \tag{34}$$

The free-energy landscape in zero field is shown in Fig. 1. At nonzero temperatures $a > 0$, the magnitude of the magnetization at the saddle is smaller than unity since it is directed perpendicularly to the easy axis, and for this orientation the "equilibrium" magnetization is smaller than in the direction along the z axis. For $a > 1$, the two saddle points, or rather the saddle circle, degenerate into a single saddle point at $n_x = n_z = 0$, and the local maximum there disappears. That is, for the magnetization to overcome the barrier, it is easier to change its magnitude than its direction. This is a phenomenon of the same kind as the phase transition in ferromagnets between the Ising-like domain walls in the vicinity of T_c (the magnetization changes its magnitude and is everywhere directed along the z axis) and the Bloch walls at lower temperatures [3, 4].

4 The Stoner-Wohlfarth curve

The Stoner-Wohlfarth curve separates the regions where there are two minima and one minimum of the free energy. On this curve the metastable minimum merges with the saddle point and loses its local stability. The corresponding condition is

$$\partial^2 f/\partial n_x^2 \times \partial^2 f/\partial n_z^2 - (\partial^2 f/\partial n_x \partial n_z)^2 = 0, \tag{35}$$

or, explicitly

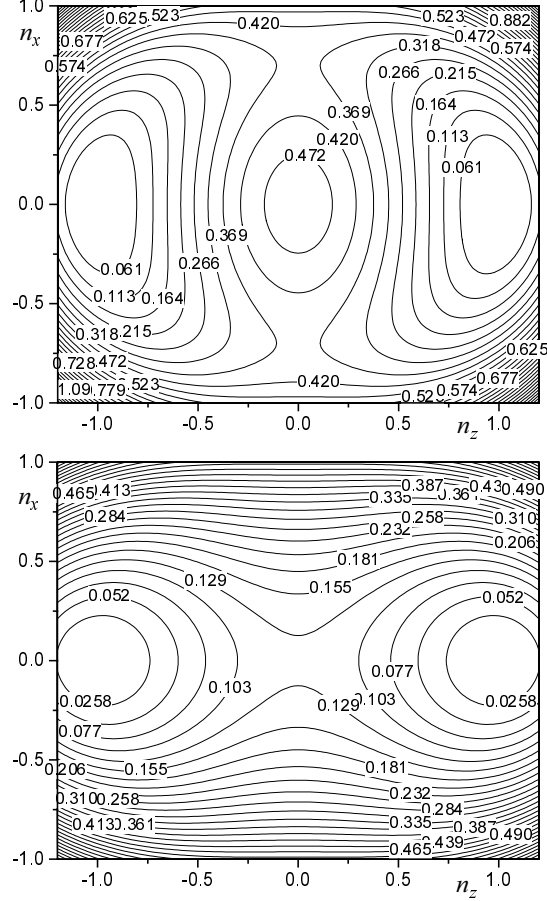$$(1 - n^2)\left(1 - 3n^2 - a\right) + 2an_z^2 = 0. \tag{36}$$

Figure 1: The free energy of a ferromagnetic particle with uniaxial anisotropy [f in Eq. (27) in zero field] for $a \equiv 2\chi_\parallel / \chi_\perp = 0.5$ (upper plot) and $a = 2$ (lower plot) corresponding to lower and higher temperatures, respectively.

Using Eq. (30), one can transform the equation above to the quartic equation for n_z

$$h_z[(2+a)n_z + 3ah_z] + 2n_z^4 = 0. \tag{37}$$

Before considering the general case, let us analyze the limiting cases $a \ll 1$ and $a \gg 1$.

At low temperatures, i.e., $a \ll 1$, the magnetization only slightly deviates from its equilibrium value, and from Eq. (36), to first order in a, one obtains

$$n^2 \cong 1 - an_z^2 \qquad \text{or} \qquad n_x^2 + (1+a)n_z^2 \cong 1. \tag{38}$$

From Eq. (37) and the analogous equation for n_x and using Eq. (30), one can derive the field dependence of n_z and n_x on the Stoner-Wohlfarth curve

$$n_z \cong -h_z^{1/3}[1 - (a/2)(h_z^{2/3} - 1/3)]$$
$$n_x \cong h_x^{1/3}[1 + (a/2)(h_x^{2/3} - 1)]. \tag{39}$$

Inserting these results in Eq. (38), one arrives at the equation for the Stoner-Wohlfarth astroid

$$h_x^{2/3} + [(1 + a/2)h_z]^{2/3} \cong 1, \qquad a \ll 1, \tag{40}$$
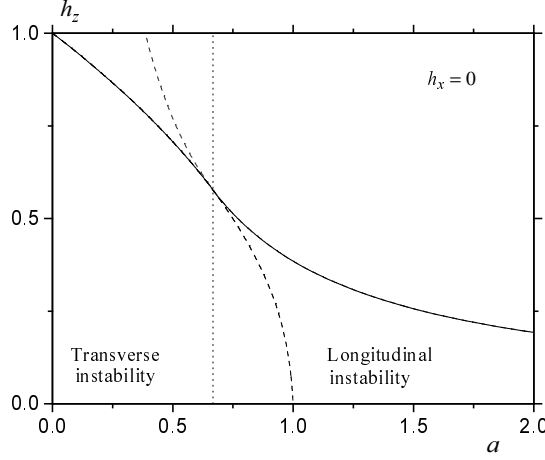
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Figure 2: Dependence $h_z(a)$ at $h_x = 0$ on the Stoner-Wohlfarth curve.

where a is given in Eq. (28). One can see that, in comparison with the standard zero-temperature Stoner-Wohlfarth astroid, i.e. at $a = 0$, h_z is rescaled. The critical field in the z direction decreases because of the field dependence of the magnetization magnitude at nonzero temperatures.

In the case $a \gg 1$, i.e. near T_c, Eq. (36) relating n_x and n_z in the equation for Stoner-Wohlfarth curve simplifies to

$$n_x^2 + 3n_z^2 \cong 1. \tag{41}$$

Using this equation together with Eqs. (30), one obtains

$$n_z \cong -(ah_z/2)^{1/3}, \qquad n_x \cong h_x. \tag{42}$$

Making use of this result in Eq. (41), one obtains another limiting case of the Stoner-Wohlfarth curve

$$h_x^2 + 3(ah_z/2)^{2/3} \cong 1, \qquad a \gg 1. \tag{43}$$

In this case, the critical field (i.e., the field on the Stoner-Wohlfarth curve) in the z direction is strongly reduced, and there is no singularity in the dependence $h_{cz}(h_x)$ at $h_x = 0$.

The qualitatively different character of the Stoner-Wohlfarth curves in these two cases is due to the different mechanisms pertaining to the loss of the local stability for the field applied along the z axis. For $h_x = 0$ the mixed derivative $(\partial^2 f/\partial n_x \partial n_z)$ in Eq. (35) vanishes, and Eq. (35) factorizes. Explicitly, we have

$$(a - 1 + n_z^2)(-1 + 3n_z^2) = 0. \tag{44}$$

Vanishing of the first factor in this equation corresponds to the loss of stability with respect to the rotation of the magnetization, $\partial^2 f/\partial n_x^2 = 0$. Vanishing of the second factor, $\partial^2 f/\partial n_z^2 = 0$, implies the loss of stability with respect to the change of the magnetization length. Using $n_x = 0$, with the help of the first equality in Eqs. (30) one obtains in the two cases

$$h_{cz} = \begin{cases} h_{z\parallel} \equiv \sqrt{1-a}, & a \leq 2/3 \\ h_{z\perp} \equiv 2/(3^{3/2}a), & a \geq 2/3. \end{cases} \tag{45}$$
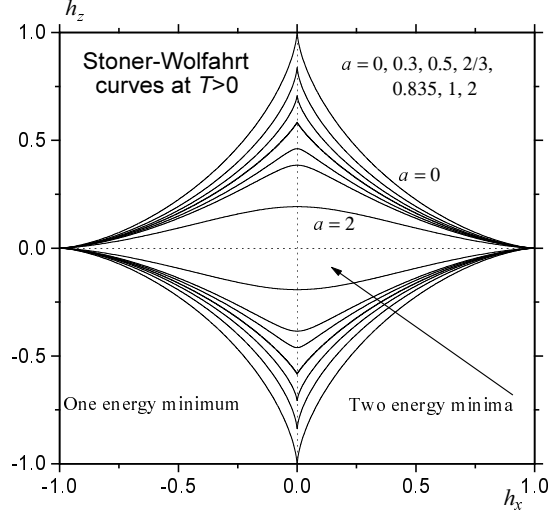
10

Figure 3: The Stoner-Wohlfarth curves at different temperatures, $a \equiv 2\chi_\parallel / \chi_\perp = 0$ $(T = 0)$, 0.3, 0.5, 2/3, 0.835, 1, 2.

Note that the transition between the two regimes occurs here at a different value of a than in Eq. (33). The dependence $h_z(a)$ at $h_x = 0$ is shown in Fig. 2.

In the general case, it is easier to find the Stoner-Wohlfarth curve numerically from Eqs. (29) and (36). The results in the whole range of a are shown in Fig. 3.

5   Hysteresis loops

In this section we use our model to study the hysteresis loop, i.e., the dependence $\mathbf{n}(\mathbf{h})$ at the stable or metastable free-energy minimum. At first we consider the case $h_x = 0$ in which the problem can be solved analytically. Setting $n_x = 0$ in the first of Eqs. (29) one obtains the cubic equation $n_z^3 - n_z - ah_z = 0$, the solution of which, for the positive branch of the hysteresis curve, reads

$$
n_z = \begin{cases}
\dfrac{2}{\sqrt{3}} \cos\left(\dfrac{\phi}{3}\right), & |h_z| \le h_{z\parallel} \\[2mm]
\left[\dfrac{ah_z}{2} + \sqrt{D}\right]^{1/3} + \left[\dfrac{ah_z}{2} - \sqrt{D}\right]^{1/3}, & |h_z| \ge h_{z\parallel},
\end{cases} \tag{46}
$$

where $h_{z\parallel}$ is given by Eq. (45) and

$$
\phi \equiv \arccos(h_z / h_{z\parallel}), \qquad D \equiv (a/2)^2 (h_z^2 - h_{z\parallel}^2). \tag{47}
$$

The negative branch of the hysteresis curve can be obtained by the reflection $h_z \Rightarrow -h_z$ and $n_z \Rightarrow -n_z$. Eq. (46) is written in the form which is explicitly real. In fact, both forms hold in the whole range $h_z \ge -h_{z\parallel}$, and there is no change of behavior at $h_z = h_{z\parallel}$, as expected on physical grounds. The characteristic values of the magnetization on the positive branch are $n_z(-h_{z\parallel}) = 1/\sqrt{3}$ and $n_z(h_{z\parallel}) = 2/\sqrt{3}$. The solution above was obtained by setting $n_x = 0$ and thus ignoring the transverse instability which can occur before the longitudinal instability at
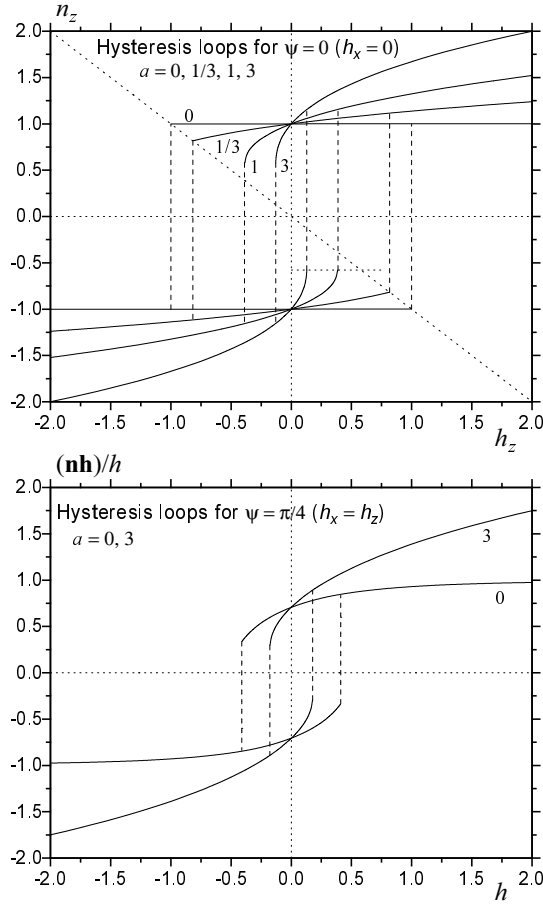
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Figure 4: Thermal effect on the static hysteresis loops $[a(0) = 0, a(T_c) = \infty]$. Upper plot: $\psi = 0$ ($\mathbf{h} \| \mathbf{e}_z$), lower plot: $\psi = \pi/4$.

$h_z = -h_{z\|}$ for the positive branch. This competition of instabilities has been studied in the previous section. It was found that for $a < 2/3$ the transverse instability occurs at the fields $|h_z| = |h_{z\|}| = \sqrt{1-a} < |h_{z\perp}|$. Thus in this case the branches of the hysteresis curves should be cut at $\pm h_{z\|}$; at these fields the system jumps to the other branch.

The analytical results obtained above for $h_x = 0$ and the numerical ones for the case of $\mathbf{h}$ directed at the angle $\psi = \pi/4$ to the easy axis are shown in Fig. 4. For $h_x = 0$, the derivative dn_z/dh_z diverges at the metastability boundary for $a \geq 2/3$ (longitudinal instability).

6 The homogeneity criterion, thermal activation, spin-waves

In the preceding sections we illustrated how the general magnetic free energy of Eq. (23) works for the simplest model of a uniaxial magnetic particle in a single-domain state at elevated temperatures. Here we discuss the possibility of observing the new types of behavior found above. This requires satisfying two rather restrictive conditions. First, the particle spends in the metastable

minimum a time that is long enough for measuring only if the barrier height energy is much larger than thermal energy. At any $T > 0$, the particle will escape from the metastable state via thermal activation with a rate exponentially small at low temperatures. For this reason, strictly speaking, the static hysteresis at nonzero temperatures does not exist and dynamic measurements are needed. At elevated temperatures, the required frequency of these measurements can become too large. Second, the free energy barrier in the single-domain state should be smaller than the barrier energy related with the formation of a domain wall which would travel through the particle and switch the magnetization from one state to the other. The two criteria can be combined as follows

$$T \ll \Delta F_{SD} < \Delta F_{DW}. \tag{48}$$

The first criterion here requires that the particle's volume is high enough whereas the second criterion requires that it does not exceed some maximal value. Let us consider, for instance, the zero field case, in which F_{SD} is the single-domain free-energy barrier of Eqs. (34) and (27). At not too high temperatures $a \lesssim 1$, one has $\Delta f \sim 1$, and the domain walls are usual Bloch walls with the energy per unit area

$$w = 2M_e^2/(q_d^2 \delta), \qquad \delta = \sqrt{\chi_\perp}/q_d, \tag{49}$$

where δ is the domain-wall width. For a spherical particle of radius R, the saddle point of the energy corresponds to the domain wall through the center of the particle. Using the definitions of parameters introduced in Sec. 2, one obtains the formula

$$\frac{\Delta F_{SD}}{\Delta F_{DW}} = \frac{Rq_d}{12\chi_\perp^{1/2}} \sim \sqrt{1 - \eta} \frac{R}{a_0}, \tag{50}$$

where a_0 is the lattice spacing. It is seen that the single-domain behavior of particles with $R \gg a_0$ requires small values of the anisotropy $1 - \eta$. Working out the first inequality in Eq. (48), one can rewrite these equations in the form

$$\left(\frac{\theta}{(1-\eta)\sigma_e^2}\right)^{1/3} \ll \frac{R}{a_0} \lesssim \frac{1}{\sqrt{1-\eta}}, \tag{51}$$

where $\theta \equiv T/T_c^{\mathrm{MFA}}$ and σ_e is the spin polarization at equilibrium. Clearly, for $\theta \ll 1$ these conditions can be satisfied. Observing the new effects exhibited by the hysteresis suggested above requires $a \equiv 2\chi_\parallel/\chi_\perp \equiv 2\bar{\chi}_\parallel/\bar{\chi}_\perp \sim 1$, which for small anisotropy, i.e. $1 - \eta \ll 1$, requires approaching T_c. Indeed, near T_c the first of Eqs. (19) yields $\bar{\chi}_\parallel \cong (2\epsilon)^{-1}$, thus $a \sim 1$ implying $\epsilon \sim 1 - \eta$. In this region in Eq. (51) one has $\theta \sim 1$ and $\sigma_e^2 \cong (5/3)\epsilon$. Then the existence of the interval for R/a_0 in Eq. (51) requires $(1 - \eta)/\epsilon \ll \epsilon$ which is impossible since in this region $(1 - \eta)/\epsilon \sim 1$ and $\epsilon \ll 1$. A similar analysis shows that also in the region where $a > 1$ Eqs. (48) cannot be satisfied.

Thus we are led to the conclusion that the qualitatively different types of behavior of single-domain magnetic particles for $a \gtrsim 1$ cannot be observed with the standard techniques. If the particle's size is small enough, the single-domain criterion is satisfied, but increasing temperature to $a \sim 1$ causes strong thermal fluctuations. The potential landscape shown in Fig. 1 is still valid but for studying

the dynamics of the magnetic particle in this range we need a special kind of Fokker-Planck equation for *non-rigid* magnetic moments. Such an equation has not been considered yet.

On the other hand, for large particle sizes the energy barriers are high and thus the process of thermal activation is suppressed, which is favorable for the observation of hysteresis loops. In this case, however, the barrier states are those with a domain wall across the particle. Analyzing these states goes beyond the scope of this article. We only mention that for $a > 1/2$ the structure of domain walls in a ferromagnet is completely different from that of a Bloch wall: The transverse magnetization component in the wall is zero everywhere whereas the longitudinal component changes its magnitude and goes through zero in the center of the wall [3]. For the observation of thermal effects in the hysteresis via inhomogeneous states, higher values of the anisotropy $1 - \eta$ are needed. In this case, thermal effects manifest themselves starting from low temperatures. An example of a strongly anisotropic material is Co, for which one obtains $1 - \eta \simeq 0.02$. This value is still much smaller than one, so that the validity condition for the magnetic free energy of Eq. (23) is satisfied.

Returning to the results of this paper, we can say that one can only observe corrections to the well-known results, such as that of Eq. (40), in the range $a \ll 1$, i.e., not close to T_c. On the other hand, one should not forget that the mean-field approximation used in this paper, while leading to qualitatively correct predictions, fails to account for some subtler effects that can be responsible for important modifications of the results. One of these effects is the influence of spin waves on the longitudinal susceptibility which enters the definition $a \equiv 2\chi_\parallel/\chi_\perp$. Whereas within the MFA $\chi_\parallel$ rapidly decreases with decreasing temperature below T_c and is independent of the anisotropy, it is *infinite* in the whole range below T_c for isotropic ferromagnets because of spin waves. The square-root singularity in the dependence $M(H)$ at zero field has been experimentally observed and reported on in Ref. [11]. In uniaxial ferromagnets, $\chi_\parallel$ becomes large for small anisotropies. This means that if the mean-field expression for a is replaced by its value taken from experiments, which is much larger due to spin-wave effects, the thermal effects discussed in this paper will considerably increase in intensity. Such a redefinition of a is, of course, not rigorously justified, although it captures the essential physics. A more involved approach taking into account spin-wave effects for an exactly solvable model confirms the concomitant increase of thermal effects on the variation of the magnetization length, as was explicitly shown for domain walls in Ref. [12].

Finally, we would like to mention that quite recently, experimental results have been obtained by Wernsdorfer et al. [13] on 3 nm cobalt nanoparticles which clearly show the disappearance of the singularity near $H_x = 0$ at a temperature circa 8 K (the blocking temperature being 14 K). The height of the experimental astroid decreases nearly as its width with increasing temperature, but it does not become flat as predicted by our calculations, which is not surprising considering the fact that $T \ll T_c$, but the disappearance of the singularity is definitive.

7 Conclusion

In this paper we have derived a macroscopic free energy for weakly anisotropic ferromagnets which is based on the mean-field approximation and is valid in the whole range of temperature interpolating between the micromagnetic energy at $T = 0$ and the Landau free energy near T_c. As an illustration, we have considered single-domain magnetic particles with uniaxial anisotropy and we have shown that thermal effects qualitatively change the free-energy landscape at sufficiently high temperatures, so that the passage from one free-energy minimum to the other is realized by the *uniform change of the magnetization length* rather than the *uniform rotation*. This also qualitatively changes the character of the Stoner-Wohlfarth curve and hysteresis loops. The latter effects cannot be observed with standard methods, however, because keeping the height of the free-energy barrier much larger than thermal energy requires so large particle sizes that the single-domain criterion is no longer satisfied. For the uniform states, the theory is valid at low temperatures, but then the thermal effects considered in the paper are small corrections to the zero-temperature results. Large thermal effects on hysteresis should be searched for at temperatures close to T_c in particles of larger sizes, where the saddle point of the free energy is an *inhomogeneous* state. Investigation of the corresponding more complicated processes is beyond the scope of this paper.

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